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Random anisotropy in UGe_2 amorphous alloy

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Abstract

Magnetic properties have been observed for the UGe_2 amorphous alloy produced by r.f. sputtering. No magnetization saturation at 50 kOe and large coercive force indicate that this alloy has marked random anisotropy. The isothermal remanence decays slowly in a manner of Arrhenius type thermal activation process. The thermomagnetic curves exhibit strong dependence on the history of cooling processes. The zero-field-cooled susceptibility $\chi_0 = M/H$ ($H = 10$ Oe), indicates that the UGe_2 amorphous alloy undergoes a higher-order phase transition to a spin-glass like phase with no spontaneous magnetization but an infinite susceptibility, as proposed by Aharony and Pytte.

Keywords: UGe_2; Amorphous alloy; Magnetism; Random anisotropy

1. Introduction

The magnetic order in amorphous alloys is dominated by two major factors: the average exchange interaction, J, and the local magnetic anisotropy, D [1]. The ferromagnetic exchange can lead to the parallel orientation of neighboring magnetic moment even in an amorphous matrix. On the other hand, the magnetic anisotropy yielded from the coupling between the orbital-angular-momentum and the local electrostatic fields tends to suppress the collinear ferromagnetism. Harris et al. assumed that the easy directions, n_i around magnetic ions are randomly distributed from site-to-site, and proposed the simple Hamiltonian (HPZ model) [2]

$$H = -J\sum_{i,j} S_i \cdot S_j - D\sum_i (n_i \cdot S_i)^2. \quad (1)$$

For transition metal amorphous alloys, the second term is usually insignificant compared with the first one. On the contrary, the second term effect is important for random anisotropy in rare-earth amorphous alloys containing non-S state ions [3]. Therefore, the magnetic response to an external magnetic field is characterized by a D/J ratio [4].

Since localized 5f electrons also have large orbital-angular-momentum, it is expected that random anisotropy plays an important role in the magnetic behavior of the uranium-based amorphous alloys. Indeed, Freitas et al. probed the random anisotropy effects into $U_{27}Fe_{73}$ amorphous alloy [5]. However this system is rather complicated because 3d electrons of iron contribute to the magnetic interactions. UGe_2 intermetallic compound is a highly anisotropic ferromagnet below $T_c = 52$ K, where the 5f electrons are rather itinerant and hybridized with the conduction electrons in this system [6]. Recently we found that the UGe_2 amorphous alloy shows magnetic order similar to the crystalline counterpart [7]. The UGe_2 amorphous alloy is a good candidate to investigate the random anisotropy effect, because the magnetism is originated only from 5f electrons.

2. Experimental

A UGe_2 target of 30 mm in diameter was prepared by arc melting of 99.9% uranium and 99.9999% germanium. A bulk specimen of the UGe_2 amorphous alloy about 100 μm thickness was deposited on a water-cooled Cu substrate by r.f. sputtering equipment. The randomness of this sample was confirmed by the X-ray diffraction measurement. After mechanical removal from the Cu substrate, detailed studies of magnetization as a function of temperature, applied magnetic field and time after switching off the

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field have been made using a SQUID magnetometer (Quantum Design MPMS).

3. Results and discussion

Initial magnetization and hysteresis loop experiments have been performed on the UGe_2 amorphous alloy at various temperatures between 5 and 60 K. The applied fields vary from 0 to 50 kOe and then from 50 to −50 kOe. In these experiments, the sample was zero-field-cooled from 90 K (a paramagnetic regime) down to the measurement temperature. Initial and reverse magnetization curves are presented in Fig. 1. A large remanent magnetization is observed at 5 and 10 K. Moreover, their initial curves have distinct inflection points at around 11 and 4 kOe, respectively, which seem to reflect the cross-over between a regime of pinning of the magnetization in metastable walls due to the random anisotropy and that of a progressive depinning due to the coupling to the applied field. Magnetization curves above 30 K show no hysteresis and their magnitudes decrease with increasing temperature. Although the low field susceptibilities show strong dependence on the temperatures, the high field ones are almost the same as each other.

The hysteresis loops for the UGe_2 amorphous alloys at some temperatures are shown in Fig. 2. The magnetization does not saturate up to 50 kOe with no temperature dependence below 10 K. The moment is the same to 0.8 μ_B/U for the polycrystalline sample in an applied field of 8.4 kOe [8] and a half of 1.43 μ_B/U for the single crystal sample along the *c*-axis (the easy direction) [9]. The shapes of these loops are symmetrical but strongly temperature dependent. In particular, both the coercive force and remanent magnetization are reduced rapidly with the increase of temperature. These behaviors have been frequently found in the rare-earth ferromagnetic amorphous alloys with random anisotropy where the magnetic pinning is dominated by thermal activation mechanism.

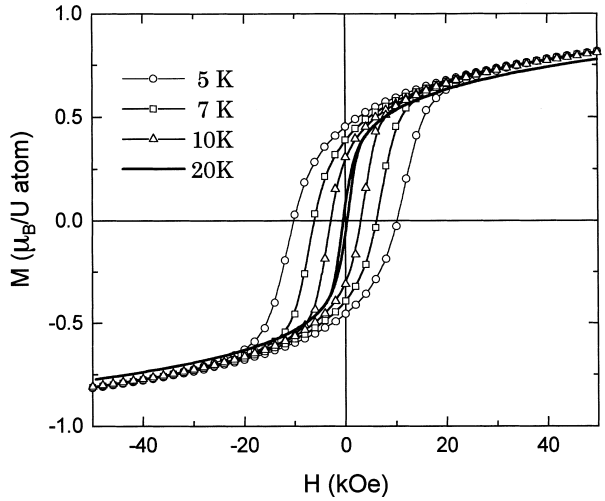

Fig. 2. Hysteresis loop of UGe_2 amorphous alloy for the temperatures below 20 K.

Magnetic after effects, known as magnetic viscosity, occurs in magnetic fields lower than the intrinsic coercive force. The time dependence of the isothermal remanent magnetization, $M_r(t)$, of the UGe_2 amorphous alloy is shown in Fig. 3. It decays slowly, approximately obeying the following relation

$$\frac{M_r(t)}{M_r(0)} = 1 - c\log t, \tag{2}$$

where $M_r(0)$ is the remanent magnetization when the applied field of 50 kOe is just taken away. A logarithmic

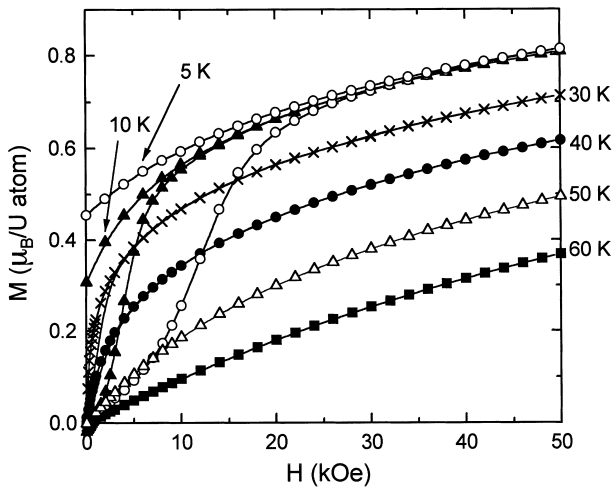

Fig. 1. Initial and reverse magnetization curves of UGe_2 amorphous alloy for the temperatures below 60 K.

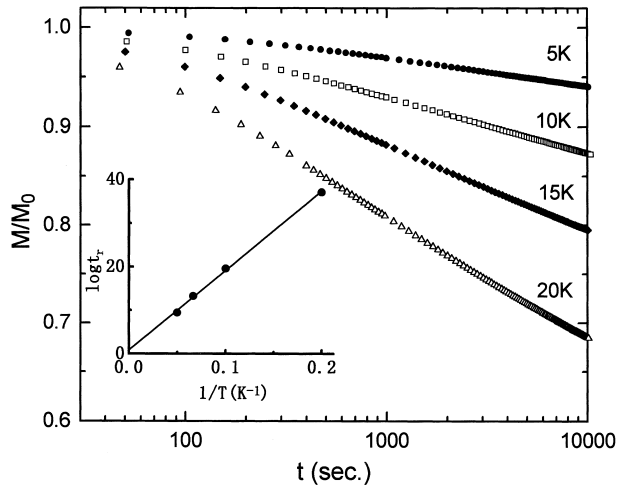

Fig. 3. Time decay of the isothermal remanence of UGe_2 amorphous alloy at various temperatures. The inset shows the temperature dependence of t_r, the extrapolated time for the remanence to decrease to zero.

dependence, typical of a thermal activation process with a wide random distribution of energy barrier, is also found in random anisotropy ferromagnets and spin glass systems. Extrapolating the curves to zero gives a relaxation time, t_r, need for the remanence to decay completely. As shown in the inset of Fig. 3, a logarithm of the decay time is proportional to the inverse of T. Therefore, the relaxation effect is obviously the Arrhenius type thermal activation process written as,

$$t_r = t_0 \exp(E_n/kT). \quad (3)$$

The activation energy in the temperature unit, E_n, about 420 K is derived from this plot, which dominates the rate-determining step.

The nature of phase transition is quite different in collinear-spin- (ferromagnetism) and non-collinear-spin-(random anisotropy or spin-glass) systems. It is theoretically predicted that a random anisotropy system undergoes a higher-order phase transition at the freezing temperature, T_f, to a spin-glass like phase with no spontaneous magnetization but an infinite susceptibility [10]. Comparison of thermomagnetic measurements between zero-field-cooled (ZFC) and field-cooled (FC) magnetization shed light on the freezing of the magnetic moments. In this procedure, the sample was initially held at 60 K for a few minutes and then cooled down to 5 K under the zero-field or the field of the measurements. The reduced magnetization, M/H, was measured with increasing temperature for several applied fields as shown in Fig. 4. There are clear thermomagnetic effects in the UGe_2 amorphous alloy: all ZFC curves have a characteristic maximum at T_f, while the FC magnetization increases gradually with decreasing temperatures even below T_f. When increasing the applied magnetic fields from 10 Oe to 5 kOe, T_f decreases from 32 to 14 K and the ZFC curves lose the sharp symmetrical cusp which is a most well-known characteristic in spin-glass like systems. Although a difference between the ZFC and FC curves is observed in ferromagnets with domain wall [11], there is no obvious domain wall in ferromagnetic amorphous alloys in general. A novel picture of the magnetic state, the correlated spin-glass (CSG), has been proposed for amorphous ferromagnets [12]. The magnetization in this state rotates smoothly over the matrix with the characteristic length because a propagation of the collinear ferromagnetic coupling is depressed by local magnetic anisotropy. It is thought that the CSG magnetic state is essentially different from the domain structure of a crystalline ferromagnet. The cusps of the ZFC curves seem to reflect the formation of the CSG state in UGe_2 amorphous alloy.

Fig. 5 shows the isothermal M/H vs. H curves obtained from Fig. 4 for the temperature above T_f. From extrapolation of the finite zero field, we can estimate the zero-field susceptibility, χ_0, which is supposed to be the same as M/H ($H=10$ Oe). It is obvious that χ_0 of the UGe_2 amorphous alloy is infinite at around T_f similar to typical spin-glass systems [13]. At T_f, we find $\chi_0 \sim 0.0015$ in a formula unit of $1/N$, where N is the demagnetization factor ($N \sim 1$). Since χ_0 does not reach the demagnetization limit, we estimate $D/J \sim 0.2$ using the relation of $\chi_0 \sim (D/J)^4$ [14].

Because a higher-order phase transition shows generally some symptoms at the critical temperature, T_c, it is worthwhile to derive the value of the critical exponent γ from the data above T_c. Assuming the usual $\chi_0 \sim (1-T/T_c)^{-\gamma}$ form, we have plotted $\chi_0/(d\chi_0/dT)$ vs. T in Fig. 6. A linear dependence was observed above 42 K with a slope of $1/\gamma$, while the susceptibility below 37 K deviates from the line and diverges at T_f. The obtained value of $\gamma=1.45$ is of the same order as sometimes observed in both collinear and non-collinear amorphous ferromagnets. However, this value is different from the mean field one of $\gamma=1$, indicating the existence of the critical fluctuation at around T_c.

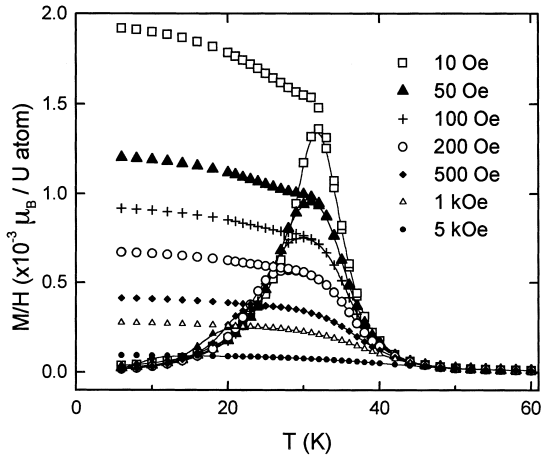

Fig. 4. Temperature dependence of ZFC (symbols with solid lines) and FC (only symbols) reduced magnetization of UGe_2 amorphous alloy for several applied fields.

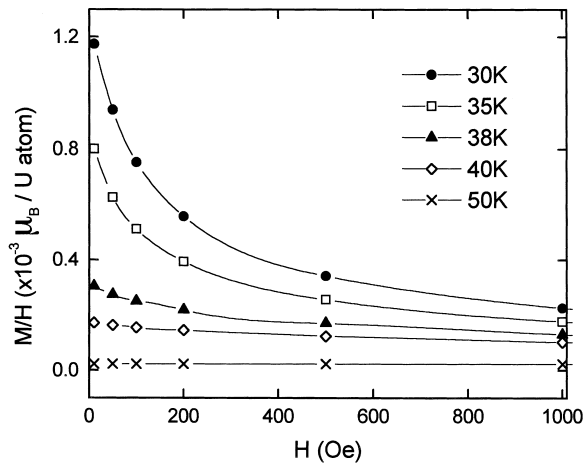

Fig. 5. Isothermal lines M/H vs. H of UGe_2 amorphous alloy for different temperatures.

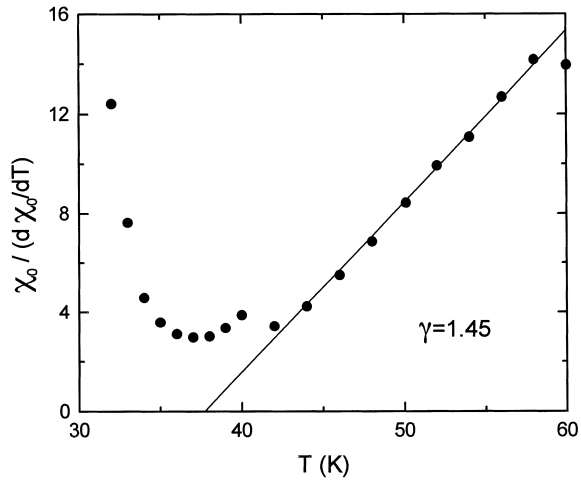

Fig. 6. Determination of γ from $\chi_0/(d\chi_0/dT) = -(T-T_c)/\gamma$ for UGe_2 amorphous alloy. M/H ($H = 10$ Oe) was adopted as χ_0.

4. Conclusion

Magnetic behavior of the UGe_2 amorphous alloy prepared by r.f. sputtering was investigated as functions of temperature, applied magnetic field and measuring time. The magnetization curves below 20 K show typical random anisotropy effects: they do not saturate in the field of 50 kOe and the value of coercive force is very large comparable to those of the non-S state rare-earth amorphous alloys. The marked time dependence is found in the magnetization induced by the external magnetic field. Thermal activation energy of 420 K is supposed to be of the order of the anisotropy energy for this alloy. The UGe_2 amorphous alloy also exhibits a distinct irreversibility in the thermomagnetic curves of ZFC and FC processes. As expected from theoretical prediction for random anisotropy systems, this alloy undergoes a higher-order phase transition to a spin-glass like phase with no spontaneous magnetization but an infinite susceptibility. A D/J ratio, obtained from the non-field susceptibility $\chi_0 \sim (D/J)^4$, of about 0.2 indicates rather strong anisotropy. Although spin correlation in this alloy is basically originated from the itinerant 5f electrons similar to the UGe_2 intermetallic compounds, it is thought that large orbital-angular-momentum of partially localized 5f electrons contributes to random anisotropy around U atoms.

Acknowledgements

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